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Muon spin relaxation study of the anomalous magnetic behavior in excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ ($0.020 \leqslant \delta \leqslant 0.101$)

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Zero-field muon-spin-relaxation measurements have revealed the hole-concentration dependence of a static magnetically ordered state of Cu spins in excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ with δ ranging from $\delta=0.020$ to 0.101. Muon-spin precession is observed at 1.7 K for $\delta \leqslant 0.0625$, consistent with the appearance of a long-range-ordered state of Cu spins. The volume fraction of the long-range-ordered state decreases rapidly with increasing δ, and a static magnetically disordered state of Cu spins becomes dominant for $\delta > 0.030$. The magnetic correlations between Cu spins are anomalously enhanced for $\delta=0.055$ and near the hole concentration is 1/8 per Cu, and the superconductivity is simultaneously suppressed. The present results confirm that the so-called 1/8 anomaly is present in the excess-oxygen-doped system in addition to substitutionally doped systems.

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I. INTRODUCTION

In the history of the high-T_c superconducting oxides, the study of the so-called 1/8 anomaly[1,2] has played an important role in the investigation of the mechanism of high-T_c superconductivity. The 1/8 anomaly is characterized by both the anomalous suppression of superconductivity and enhancement of the magnetic correlation between Cu spins around the hole concentration of 1/8 per Cu. After the launch of the so-called stripe model of spins and holes,[3] the 1/8 anomaly has been regarded as being due to the static stabilization of dynamically fluctuating stripes and the anomaly has been studied renewedly in order to understand the stripe model.

The 1/8 anomaly was first discovered in $La_{2-x}Ba_xCuO_4$ (LBCO) (Refs. 1 and 2) and seen also in $La_{2-x}Sr_xCuO_4$ (LSCO) (Refs. 4–6). The 1/8 anomaly has also been confirmed from the zero-field muon-spin-relaxation (ZF-μSR) and transport measurements in the partially Zn-substituted systems $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ (Refs. 7–10) and $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ (Ref. 11) whose crystal structures are different from those of LBCO and LSCO, apart from the inclusion of the two-dimensional CuO_2 planes. Moreover, it has been revealed from the transport measurements on $La_{2-y-x}Ce_yBa_xCuO_4$ ($y=0$ and 0.02) (Ref. 12) and $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ ($x=0$ and 0.2) (Ref. 13) that the hole concentration of 1/8 per Cu is essential to the 1/8 anomaly as its name implies. Accordingly, the 1/8 anomaly has been suggested to be an electronic phenomenon common to all hole-doped superconducting oxides with the CuO_2 plane.

With regard to testing of the universality of the 1/8 anomaly, it is of interest to study the electronic properties of excess-oxygen-doped $La_2CuO_{4+\delta}$ (LCO), because this system provides another example of hole-doped superconducting oxides with the CuO_2 plane.[14,15] The hole concentration of excess-oxygen-doped LCO is controlled by the amount of the excessively doped oxygen which is located at an interstitial site between CuO_2 planes.[16] Because of phase separation of the excess oxygen into oxygen-rich and oxygen-poor regions,[17] however, the relationship between the magnetically ordered and superconducting states in LCO has previously been investigated only at a few specific excess-oxygen concentrations.[17–22]

In order to achieve continuous control of the hole concentration, Koike *et al.*[23] and Mikuni *et al.*[24] synthesized a series of samples of excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ (LNCO). In these samples, on account of the random potential introduced in the LaO plane by the partial substitution of Nd^{3+} for La^{3+}, the phase separation of the excess oxygen is suppressed and the hole concentration per Cu corresponds uniformly to 2δ (Refs. 25–27). These authors succeeded in obtaining the detailed hole-concentration dependence of the superconducting transition temperature T_c against continuously changing δ and found anomalous suppression of T_c around $\delta=0.0625$ where the hole concentration is 1/8 per Cu. In these studies, μSR measurements were carried out on excess-oxygen-doped LNCO with $\delta = 0.0625$ at the RIKEN-RAL Muon Facility in the UK using a pulsed positive muon beam, and the appearance of a static magnetically ordered state of Cu spins was found at temperatures below about 40 K. These results strongly suggest the existence of the 1/8 anomaly in the excess-oxygen-doped LNCO system as well.

However, in these previous studies of excess-oxygen-doped LNCO,[23,24] the detailed hole-concentration dependence of the magnetically ordered state has not been obtained. Therefore, we have expanded the earlier experiment on LNCO to cover a wide range of δ from 0.020 up to 0.101 and have focused on variations of the magnetically ordered state in order to confirm the existence of the 1/8 anomaly from the ZF-μSR measurements.

II. EXPERIMENT

Polycrystalline samples of excess-oxygen-doped LNCO were prepared by the usual solid-state-reaction method. The content of the excess oxygen in each sample was controlled by the electrochemical oxidation method. Details of the sample preparation and the content of the excess oxygen are described in a separate paper.[24]

In our previous μSR measurement by Mikuni *et al.*[24] on the static magnetically ordered state of excess-oxygen-doped LNCO with $\delta = 0.0625$, information on the muon-spin polarization in a very-early-time region was not available and no clear muon-spin precession was observed even at 0.3 K due to the depolarization being faster than the time resolution of the pulsed muon beam. Thus, in order to obtain accurate information of the muon-spin depolarization behavior in the early-time region, ZF-μSR measurements have been carried out at the Paul Scherrer Institute (PSI) in Switzerland using a continuous muon beam. The time resolution available at PSI is 0.625 nsec and is about 32 times better than that at the RIKEN-RAL Muon Facility.

Each sample consisted of a disk pellet with a diameter of about 10 mm which was fixed onto a high-quality silver plate with Apiezon-N grease and put in a gas-flow-type cryostat to be cooled down to 1.7 K. Spin-polarized positive surface muons were injected into the sample. The direction of the spin polarization was parallel to the beamline. The μSR time spectrum—namely, the time evolution of the asymmetry parameter of the muon-spin polarization, $A(t)$—was obtained from the ratio of numbers of muon events counted by the forward and backward counters, which were aligned on the upstream and downstream sides to the sample, respectively. That asymmetry parameter is defined as $A(t) = [F(t) - \alpha B(t)]/[F(t) + \alpha B(t)]$, where $F(t)$ and $B(t)$ are numbers of the total muon events detected by the forward and backward counters at a time t, respectively. The parameter α is a calibration factor reflecting the relative counting efficiencies between the forward and backward counters. All time spectra have been corrected by α during the analysis procedures. The asymmetry at $t=0$, $A(0)$, is the initial asymmetry.

III. RESULTS

Figure 1 shows the typical ZF-μSR time spectra obtained at various temperatures in excess-oxygen-doped LNCO with

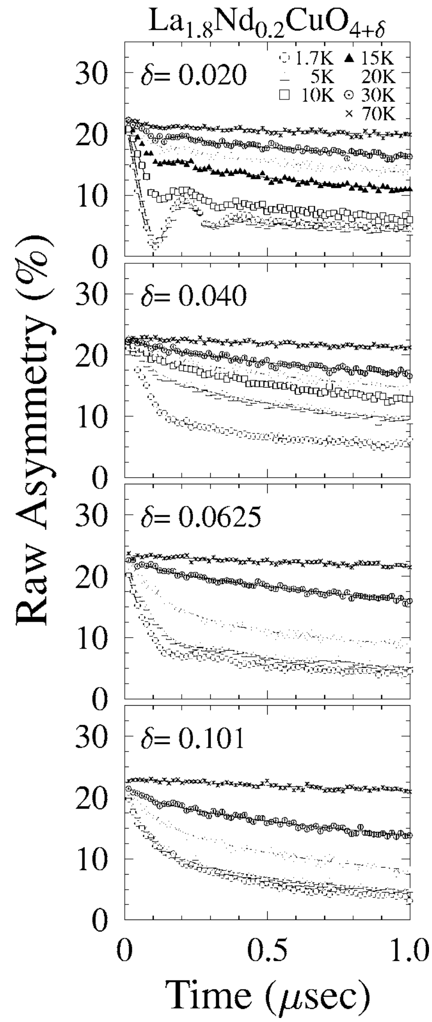

FIG. 1. Typical ZF-μSR time spectra of excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ with $\delta = 0.020$, 0.040, 0.0625, and 0.101 at various temperatures. The hole concentration of the sample with $\delta = 0.0625$ is 1/8 per Cu. Solid lines are the best-fit result using the multicomponent function of $A(t) = A_0 e^{-\lambda_0 t} + A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} \cos(\omega t + \phi)$.

$\delta = 0.020$, 0.040, 0.0625, and 0.101. The time spectrum of the sample with $\delta = 0.020$ at 70 K seems to show some oscillation. The amplitude of the oscillation becomes large with decreasing temperature, so that the muon-spin precession is clearly seen at 1.7 K. The observation of the muon-spin precession indicates a well-ordered magnetic state and strongly suggests the formation of a long-range-ordered state of Cu spins. As for the other samples, the time spectrum at 70 K is nearly flat. This means that muons slowly depolarize by nuclear dipole fields distributed at the muon site; that is, Cu spins are fluctuating on a much shorter time scale than the μSR time window (10^{-6}–10^{-11} sec). The time spectrum of each sample changes with decreasing temperature, and fast depolarization behavior starts to appear. Figure 2 displays time spectra at 1.7 K of all measured samples. Clear muon-spin precession is observed up to $\delta = 0.03$ but is quite difficult to be seen at $\delta = 0.04$ and 0.055. The muon-spin precession with tiny amplitude can be seen again at $\delta = 0.0625$ and is no longer observed for $\delta \geqslant 0.080$.

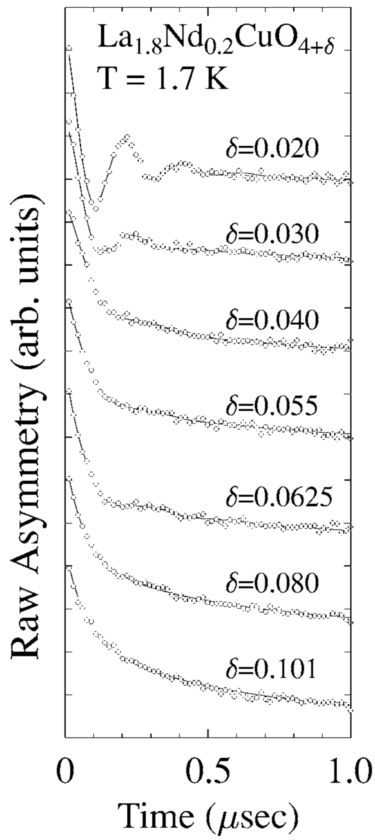

FIG. 2. ZF-μSR time spectra of excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ at 1.7 K. Solid lines are the best-fit results using the multicomponent function of $A(t)=A_0e^{-\lambda_0 t}+A_1e^{-\lambda_1 t}+A_2e^{-\lambda_2 t}\cos(\omega t+\phi)$.

Figure 3 shows time spectra in some longitudinal fields (LF's) up to 2 kG obtained in excess-oxygen-doped LNCO with $\delta=0.0625$ at 1.7 K. A LF was applied in the same direction of the muon beamline which is parallel to the muon-spin direction. The time spectrum which shows the depolarization behavior in ZF recovers with increasing LF. Long-time depolarization remains in each field, and the time spectrum does not change at all above 1 kG. The existence of long-time depolarization indicates the existence of a strongly fluctuating internal field at the muon site accomodating the static internal field caused by the long-range-ordered state. Taking into account that Nd moments in high-T_c oxides are dynamically fluctuating causing the appearance of strongly fluctuating internal fields at the muon site,[28,29] the result of the LF dependence of the time spectrum shown in Fig. 3 suggests that Nd moments are still dynamically flucating, producing the dynamic depolarization behavior of the muon spin in the current system.

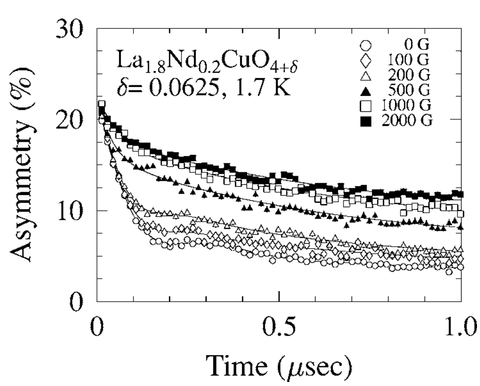

FIG. 3. LF-μSR time spectra of excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ with $\delta=0.0625$ at 1.7 K. Solid lines are the best-fit results using the multicomponent function of $A(t)=A_0e^{-\lambda_0 t}+A_1e^{-\lambda_1 t}+A_2e^{-\lambda_2 t}\cos(\omega t+\phi)$.

In order to analyze time spectra, the following multicomponent function was used as carried out in our previous paper:[24]

$$A(t) = A_0e^{-\lambda_0 t} + A_1e^{-\lambda_1 t} + A_2e^{-\lambda_2 t}\cos(\omega t + \phi). \quad (3.1)$$

The first term approximates the slowly depolarizing component due to the nuclear dipole fields distributed at the muon site[30] and also probably due to the dynamically fluctuating internal field caused by dynamically fluctuating Nd moments. The second and third terms show, respectively, the fast depolarizing component and the muon-spin precession component due to the appearance of the long-range-ordered state at low temperatures. The parameters A_0, A_1, and A_2 are the initial asymmetries at $t=0$, and λ_0, λ_1, and λ_2 are the depolarization rates of each component. The parameters A_2 and λ_2 correspond to the amplitude at $t=0$ and damping rate of the muon-spin precession, respectively. The parameters ω and ϕ are the frequency and phase of the muon-spin precession. The solid lines in Figs. 1 and 2 show the best-fit results using Eq. (3.1). The use of a multicomponents function to analyze time spectra assumes the appearance of some magnetic domain structures in the sample. The sum of the initial asymmetries of each sample—that is, the total initial asymmetry $A_0+A_1+A_2$—was estimated from the best fit of the time spectrum obtained at temperatures below 5 K. This total asymmetry was fixed when the other time spectra at different temperatures were analyzed.[31]

Figure 4 shows the temperature dependences of A_0, A_1, and A_2 in excess-oxygen-doped LNCO with $\delta=0.020$, 0.040, 0.0625, and 0.101. It is found that A_0 decreases with decreasing temperature for all samples. The decrease of A_0 is regarded as resulting from the magnetic transition—namely, the growth of a static magnetically ordered state of Cu spins as observed in single crystals of excess-oxygen-doped LCO.[6,8,9,18–20,22,24,34] In the case of $\delta=0.040$, the decrease of A_0 tends to start at lower temperature compared to other samples. This means that the appearance of the magnetically ordered state shifts to the lower-temperature side at $\delta=0.040$.

The saturated value of A_0 for $\delta\leqslant 0.0625$ is nearly one-quarter of the total initial asymmetry, which is a little smaller than the theoretically expected value—i.e., 1/3 (Ref. 30). This might be due to the influence of dynamically fluctuating Nd moments or due to poor estimations of the zero level of the time spectrum.[31]

In contrast to the decrease of A_0 with decreasing temperature, A_1 increases with decreasing temperature, reflecting the growth of the static magnetically ordered state. As for A_2, its value for $\delta=0.020$ is nonzero even at 70 K, indicating the presence of some remnant volume fraction of the long-range-

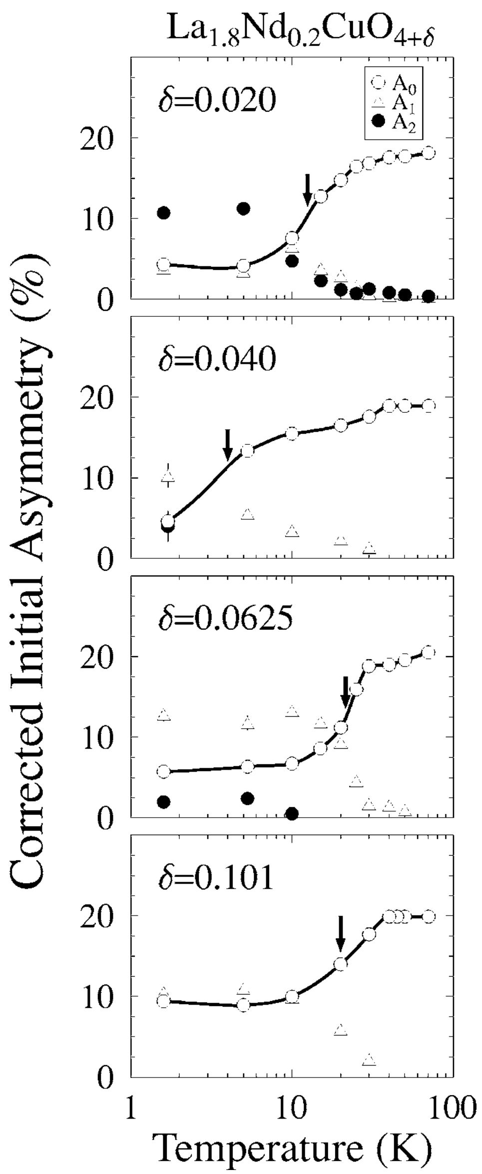

FIG. 4. Temperature dependences of the corrected initial asymmetries of the slowly depolarizing component A_0, the fast depolarizing component A_1, and the precession component A_2, respectively, for δ=0.020, 0.040, 0.0625, and 0.101 in $La_{1.8}Nd_{0.2}CuO_{4+\delta}$. Arrows indicate the characteristic transition temperature T_m^{μ}, defined at the midpoint of the change of A_0. Solid lines are guides to the eye.

ordered state of Cu spins. The value A_2 for δ=0.020 gradually increases with decreasing temperature below 70 K and is saturated at temperatures below about 5 K. For δ=0.040 and 0.055, it is hard to see the muon-spin precession even at 1.7 K. However, taking into account that the nonsmooth curve of the time spectrum around 0.15 μsec shown in Fig. 2 cannot be well described by a two-exponential components function (A_2=0), it is concluded that there exists a small amount of the muon-spin precession component at 1.7 K for δ=0.040 and 0.055. In the case of δ=0.0625, on the other hand, A_2 can be determined at temperatures below 10 K and is saturated below 5 K with a smaller value than that for δ =0.020.

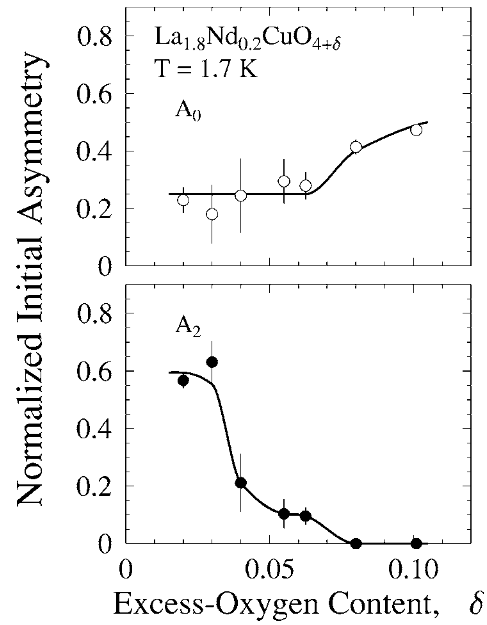

FIG. 5. Excess-oxygen-content dependence: namely, the hole concentration dependence of the normalized initial asymmetries of A_0 and A_2 at 1.7 K in excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$. Solid lines are guides to the eye.

Figure 5 shows the excess-oxygen-content dependences: namely, the hole-concentration dependences of the normalized initial asymmetries of A_0 and A_2 at 1.7 K. Values of A_0 and A_2 are normalized by the total initial asymmetry. It is found that the value of A_0 is roughly constant for $0.020 \leqslant \delta \leqslant 0.0625$, indicating that almost all Cu spins are statically stabilized at 1.7 K. For $\delta \geqslant 0.080$, on the other hand, A_0 increases with increasing δ. The value of A_2 decreases rapidly with increasing δ for $\delta > 0.030$ and seems to show a plateau or a small local peak around δ=0.0625. The A_2 component can no longer be determined for $\delta \geqslant 0.080$ because of the disappearance of the muon-spin precession.

Figure 6 displays the temperature dependence of the internal field at the muon site, $\mathbf{H}_{int}$, which is coming from Cu spins in the long-range-ordered state, for δ=0.020, 0.030, and 0.0625. Values of $\mathbf{H}_{int}$ were calculated from ω using the equation of $\mathbf{H}_{int}=\omega/\gamma_{\mu}$, where γ_{μ} is the gyromagnetic ratio of the muon (γ_{μ}=13.55 MHz/kOe). It is commonly found

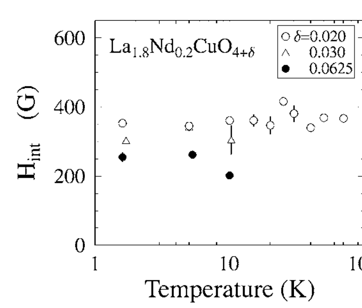

FIG. 6. Temperature dependence of the internal field at the muon site, $\mathbf{H}_{int}$, obtained from the analysis of the muon-spin precession component for δ=0.020, 0.030, and 0.0625 in $La_{1.8}Nd_{0.2}CuO_{4+\delta}$.

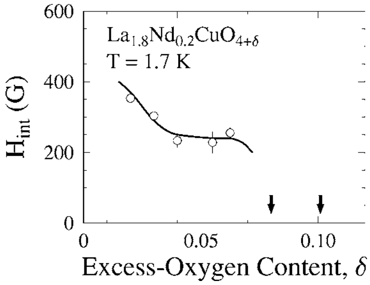

FIG. 7. Excess-oxygen content dependence: namely, the hole-concentration dependence of the internal field at the muon site, $\mathbf{H}_{int}$, at 1.7 K in excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$. Arrows mean that no meaningful value can be determined because of the disappearance of the muon-spin precession. The solid line is a guide to the eye.

that $\mathbf{H}_{int}$ is almost independent of temperature, while the precession amplitude depends upon temperature as shown in Fig. 4. The hole-concentration dependence of $\mathbf{H}_{int}$ at 1.7 K is displayed in Fig. 7. The $\mathbf{H}_{int}$ for δ=0.020 is about 15% smaller than 420 G in nondoped LCO with δ=0 (Ref. 32). The $\mathbf{H}_{int}$ gradually decreases with increasing δ and shows a plateau in its δ dependence around δ=0.0625 as well as the δ dependence of A_2. A simple estimate of the magnetic moment of Cu spins can be obtained from the comparison with $\mathbf{H}_{int}$ observed in nondoped LCO with δ=0. The value obtained is about $0.3\mu_B$ for δ=0.0625. It is of interest that this value is the same as that of LSCO (Ref. 6) and LBCO (Ref. 33) at a hole concentration of about 1/8 per Cu.

As for the muon-spin depolarization rates, all of λ_0, λ_1, and λ_2 are independent of temperature at low temperatures below about 40 K for each sample. This indicates that only volume fractions of the three magnetically classified domains (areas described by A_0, A_1, and A_2) change at low temperatures. Figure 8 shows the hole-concentration dependence of λ_2 obtained at 1.7 K. The value of λ_2 increases with increasing δ. After showing a maximum at δ=0.030, λ_2 decreases with increasing δ. This suggests that the magnetic order for $\delta<0.030$ is different from that for $\delta>0.030$. As for δ=0.040 and 0.055, it is guessed that the muon-spin precession component smears out because of the fast depolarization rate of λ_2 and the small value A_2 so as to become hard to be seen clearly in the time spectrum at 1.7 K.

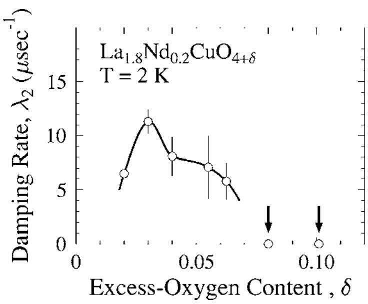

FIG. 8. Excess-oxygen-content dependence: namely, the hole-concentration dependence of the depolarization rate of the muon-spin precession component λ_2 at 1.7 K in excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$. Arrows mean that no meaningful value can be determined because of the disappearance of the muon-spin precession. The solid line is a guide to the eye.

IV. DISCUSSION

The ratios of A_0, A_1, and A_2 to the total initial asymmetry are in rough correspondence to the volume fractions of the respective components. Accordingly, the value of $A_2/[(A_0+A_1+A_2)-A_{0_{min}}]$ is regarded as the volume fraction of the long-range-ordered state,[34] where $A_{0_{min}}$ is the saturated value of A_0 at 1.7 K. The $A_{0_{min}}$ is estimated from the result shown in the upper panel of Fig. 5 to be one-quarter of A_0. Accordingly, it is estimated that about 80% of Cu spins form the long-range-ordered state for δ=0.020 and 0.030. The rapid reduction of A_2 with increasing δ shown in Fig. 5 means that the long-range-ordered state is strongly suppressed by increasing hole concentration. On the other hand, A_0 remains saturated at its minimum value up to δ=0.0625, showing that almost all Cu spins are statistically frozen within the μSR time window. These facts indicate that the volume fraction of the static magnetically disordered state of Cu spins increases with increasing hole concentration for $\delta>0.030$ at the expense of the long-range-ordered one.

As seen in Fig. 4, the magnetic transition which is estimated from the change of A_0 versus temperature is found to be broad for all samples. The transition width is about 20 K (from 30 K to 10 K) for δ=0.0625 and is larger than that observed in LSCO with a similar hole concentration. For instance, the transition width for LSCO with x=0.115 is about 5 K (Ref. 6). In addition, as shown in Fig. 6, $\mathbf{H}_{int}$ is independent of temperature even though A_2 varies with temperature. These results remind us of an inhomogeneous nature—that is, a gradual expansion of static magnetic domains with decreasing temperature, as mentioned above. The increase of A_0 for $\delta\geqslant 0.08$ indicates that the growth of magnetic domains has stopped at temperatures below 10 K. Therefore, there remain two phases even at 1.7 K for $\delta \geqslant 0.08$: the static disordered phase and a phase where Cu spins fluctuate at rates beyond the μSR time window. In the case of δ=0.101, the fraction of the static disordered phase shrinks to be about two-third at 1.7 K. The same tendency of both the broad change in the temperature dependence of A_0 and the temperature independence of $\mathbf{H}_{int}$ has also been reported in excess-oxygen-doped LCO with δ=0.11 (Ref. 22). The inhomogeneous nature may be due to the microscopic inhomogeneity of the excess oxygen, but the true origin is not cleat at present.

Although the present μSR results have revealed the likely inhomogeneous nature of excess-oxygen-doped LNCO, the dependence on the excess-oxygen content of a characteristic transition temperature T_m^{μ}, defined as the midpoint of the change of A_0 and shown by an arrow in Fig. 4, may be meaningful. In the current case, the defined T_m^{μ} means a sort of average of the inhomogeneous transitions of magnetic domains. All T_m^{μ}'s are shown by arrows in Fig. 4 and summarized in Fig. 9 together with T_c of each sample determined from the resistivity measurements by Mikuni *et al.*[24] The error bar is defined to be about ±10% of the value of the midpoint of the temperature dependence of the initial asym-

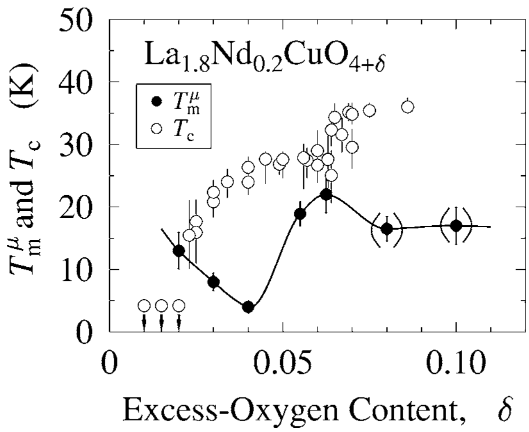

FIG. 9. Dependences on the excess-oxygen content of T_m^μ (solid circles) and T_c (open circles) (Ref. 24). The data points for $\delta \geqslant 0.080$ cannot be compared with other data points directly because there remain two phases of the static disordered state and a phase where Cu spins fluctuate beyond the μSR time window ($10^{-6}-10^{-11}$ sec). Accordingly, the data points at δ=0.080 and 0.101 are surrounded by brackets. Solid line is guide to the eye.

metry. The T_m^μ is about 13 K for δ=0.020 and decreases gradually with increasing δ. After showing a minimum at δ =0.040, T_m^μ starts to increase with increasing δ and exhibits a peak at δ=0.0625 where the hole concentration is 1/8 per Cu. For $\delta > 0.0625$, T_m^μ tends to decease and seems to saturate for $\delta \geqslant 0.080$.

The decrease of T_m^μ with increasing δ for $\delta > 0.030$ indicates the destruction of the magnetic correlations between Cu spins and is simply explained as being a result of the local frustration effect induced by doped holes in the antiferromagnetic spin arrangement as in the case of LSCO in the underdoped region.[35] The remarkable point is the appearance of the maximum of T_m^μ at δ=0.0625 where a dip of T_c is observed. The appearance of the local maximum of T_m^μ shows the enhancement of the magnetic correlation between Cu spins. Both the anomalous suppression of superconductivity and the enhancement of the magnetic correlations around the hole concentration of 1/8 per Cu are the typical behavior associated with the 1/8 anomaly.[8,9,11] Moreover, the hole-concentration dependences of both $\mathbf{H}_{int}$ and the volume fraction of the long-range-ordered state exhibit plateaus around δ=0.0625, as displayed in Figs. 5 and 7, supporting the anomalous enhancement of the magnetic correlations between Cu spins at δ=0.0625. Therefore, the present result shows that the 1/8 anomaly exists in excess-oxygen-doped LNCO also.

Taking into account the result that $\mathbf{H}_{int}$ for δ=0.0625 is nearly equal to those of LSCO and LBCO at a hole concentration of about 1/8 per Cu, it is suggested that in LNCO at δ=0.0625, there appears a Cu-spin arrangement similar to that of LSCO and LBCO. Accordingly, it is guessed for δ=0.0625 that the static ordered stripes of spins and holes are formed in the region described by A_2 and that static disordered stripes are formed in the region described by A_1. What is of particular interest is whether the magnetically ordered state and the superconducting state coexist in each sample or not, since bulk superconductivity has been confirmed by the susceptibility measurements in excess-oxygen-doped LNCO for $\delta \geqslant 0.03$ (Refs. 23 and 24). More detailed work is necessary for a clarification of this question.

V. SUMMARY

ZF-μSR measurements have been carried out on excess-oxygen-doped $La_{1.8}Nd_{0.2}CuO_{4+\delta}$ over a wide range of δ from 0.020 to 0.101 in order to study the variation of the magnetic correlations between Cu spins against the hole concentration.

Muon-spin precession is observed at 1.7 K in the samples for $\delta \leqslant 0.0625$, indicating the appearance of a long-range-ordered state of Cu spins. The precession frequency of the muon spin is independent of temperature, while the precession amplitude depends on temperature. The volume fraction of the long-range-ordered state decreases rapidly with increasing δ for $\delta > 0.030$, being replaced by a static magnetically disordered state which increases alternatively. For $\delta \geqslant 0.080$, there remain two phases consisting of the static disordered phase and a phase where the Cu-spin fluctuation is fast compared to the μSR time window. The magnetic transition in each sample is found to be broader than that of LSCO. These results are explained as being the result of the gradual development of static magnetic domains with decreasing temperature, suggesting an inhomogeneous nature of LNCO.

The magnetic correlations between Cu spins are suppressed with increasing δ for $\delta > 0.030$ and are anomalously enhanced for δ=0.055 and show a peak at δ=0.0625 where the hole concentration is 1/8 per Cu. Taking into account the result that the superconductivity is also anomalously suppressed around δ=0.0625, the present study provides strong evidence that the so-called 1/8 anomaly exists in also the excess-oxygen-doped LNCO system.

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